

## An evaluation of activated bismuth isotopes in environmental samples from the former Western Pacific Proving Grounds

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$^{207}\text{Bi}$  ( $T_{1/2} = 32.2$  y) was generated by activation of weapons material during a few “clean” nuclear tests at the U.S. Western Pacific Proving Grounds of Enewetak and Bikini Atolls. The radionuclides first appeared in the Enewetak environment during 1958 and in the environment of Bikini during 1956. Crater sediments from Bikini with high levels of  $^{207}\text{Bi}$  were analyzed by gamma-spectrometry in an attempt to determine the relative concentrations of  $^{208}\text{Bi}$  ( $T_{1/2} = 3.68 \cdot 10^5$  y). The bismuth isotopes were probably generated during the “clean”, 9.3 Mt Poplar test held on 7/12/58. The atom ratio of  $^{208}\text{Bi}$  to  $^{207}\text{Bi}$  ( $R$  value) ranges from -12 to over 200 in sections of core sediments from the largest nuclear crater at Bikini atoll. The presence of bismuth in the device is suggested to account for  $R$  values in excess of 10.

### Introduction

Radioactive fission and fusion products (primarily  $^3\text{H}$ ) unspent nuclear fuel and activation products were produced during the United States nuclear test program at the Western Pacific Proving Grounds (PPG) between 1946 and 1958. A fraction of the radioactivity resulting from the exploded devices was deposited to the local environments of the test site atolls, Bikini and Enewetak.

The activation products were produced from weapon components or on-site materials through reactions involving neutrons or possibly charged particles generated during the nuclear explosions. One of the many activation products eventually identified at the test sites was  $^{207}\text{Bi}$  ( $T_{1/2} = 32.2$  y).<sup>1</sup> LOWMAN and PALUMBO<sup>2</sup> first reported the presence of the radionuclide in algae collections made at Enewetak in 1960 after separating bismuth from the samples, and identifying  $^{207}\text{Bi}$  from its principal gamma-rays ( $E_\gamma = 570$  keV 100%; 1064 keV 75.8%; 1.770 keV 7.0%)<sup>1</sup> using a NaI detector. The radionuclide was later reported in different environmental samples at Bikini atoll, as well as at Enewetak, during a radiological survey<sup>3,4</sup> of the two atolls conducted in 1964. Subsequently concentrations of  $^{207}\text{Bi}$  have been reported in a variety of different samples, including many dietary items, from the Marshall Island and have been discussed to date, in over 25 publications or reports. Only those reports pertinent to this paper are referenced. Detailed dose assessments, that include  $^{207}\text{Bi}$  in the entire marine food chain, have been made for Bikini and Enewetak.<sup>5</sup>

Low level fallout amounts of the radionuclide also has been reported in samples from the Arctic<sup>6</sup> and Antarctic<sup>7</sup> and from locations in the United States<sup>8–11</sup> and Japan.<sup>12–19</sup> Its somewhat infrequent detection in samples from the northern hemisphere is surprising since it was reported with fallout radionuclides detected in ground level air in the U.S. Pacific Northwest<sup>11</sup> and Denmark<sup>6</sup> for extended periods between 1961 and 1966.

The radionuclide has also been reported as an impurity in crystals, in purified chemicals and some raw materials.<sup>19–21</sup> It was suggested that the large 58 Mt thermonuclear test by the former USSR held on 30 Oct 1961 generated most of the fallout  $^{207}\text{Bi}$  found in samples from the northern hemisphere.<sup>6</sup> This was a relatively “clean” device with the largest fusion yield of any atmospheric test.

The presence of  $^{207}\text{Bi}$  in the environment clearly shows that there was one or possibly more than one reaction responsible for forming the radionuclide during the history of nuclear testing at the PPG and elsewhere. The reactions and conditions by which the radionuclide is produced in the tests have not yet been clearly defined. Some investigators<sup>2,11,21</sup> suggest that the production of  $^{207}\text{Bi}$  was by a  $^{209}\text{Bi}(n,3n)$  reaction. Stable bismuth was therefore required to be associated with the devices. Others<sup>6,9,20–22</sup> propose that the radionuclide resulted during high yield thermonuclear tests from charged particle (p, d, t, etc.) reactions with isotopes of lead assumed to be component parts of the weapon. There is support for the latter suggestion since it was stated<sup>23,24</sup> that the large Mt explosion of 30 Oct. 1961 contained lead. It was possible that activation of this lead was responsible, in some part, for the fallout  $^{207}\text{Bi}$  measured in the samples reported from the Northern Hemisphere. However, it will be shown that the presence of lead in or near thermonuclear devices is by no means a guarantee that  $^{207}\text{Bi}$  will be produced by activation.

In tests where  $^{207}\text{Bi}$  was produced there also could have been a comparable or greater number of atoms of  $^{208}\text{Bi}$  ( $T_{1/2} = 3.68 \cdot 10^5$  y)<sup>1</sup> formed by neutron or charged particle activation. Neutron cross section data highly favors formation of  $^{208}\text{Bi}$  (threshold-7.5 MeV)<sup>1</sup> over  $^{207}\text{Bi}$  (threshold-14.35 MeV)<sup>1</sup> in  $^{209}\text{Bi}(n,xn)$  reactions below 19.5 MeV.<sup>1</sup> Long term irradiation of bismuth in the fast neutron flux of a reactor yielded both isotopes.<sup>25</sup> The cross sections for fission neutrons were 9.3 mb and 18  $\mu\text{b}$ , respectively, for the (n,2n) and (n,3n) reactions.<sup>25</sup>

Because of the extremely small cross section it is unlikely that much of the  $^{207}\text{Bi}$  inventory found in the PPG could have resulted from activation of bismuth with fission spectrum neutrons.

Neutrons resulting directly from  $^2\text{H}$ – $^3\text{H}$  fusion in a thermonuclear device will attain a maximum energy of 14.1 MeV. Neutrons with energies between 8 and 14.1 MeV are capable of activating bismuth to produce  $^{208}\text{Bi}$  but not  $^{207}\text{Bi}$ . It would be possible for  $^{208}\text{Bi}$  to be present in the environment independent of any  $^{207}\text{Bi}$ . However, more ( $>14.1$  MeV) energetic neutrons can be generated in a thermonuclear explosion but this requires additional reactions involving high-energy projectiles. SHINOHARA et al.<sup>21</sup> suggest one possibility when LiD is present as a fuel in a fusion device. Tritons will be generated with energies above 2.7 MeV from the  $^6\text{Li}(n,\alpha)^3\text{H}$  reaction. If these energetic tritons fuse with the deuterium fuel, neutrons will be produced with energies between 10.8 and 19.7 MeV at a maximum. These high energy neutrons have the potential to initiate (n,2n) and (n,3n) reactions with stable bismuth.

There are a number of reactions involving charged particles and lead isotopes that could be responsible for generating both  $^{207}\text{Bi}$  and  $^{208}\text{Bi}$ . YOSHIMURA et al.<sup>19</sup> argue that the  $^{206,207}\text{Pb}(d,xn)$  reactions are more important than others in forming  $^{207}\text{Bi}$  from lead components in a thermonuclear device. The same case could be made for the formation of  $^{208}\text{Bi}$  from  $^{207,208}\text{Pb}(d,xn)$  reactions.

There has been no published information of the concentrations of  $^{208}\text{Bi}$  at the PPG. However, there has been one reported attempt to measure “environmental  $^{208}\text{Bi}$  levels” in a sample of bismuth nitrate salt processed in Japan during 1984.<sup>21</sup> The  $^{208}\text{Bi}$  to  $^{207}\text{Bi}$  atom ratio in this sample was estimated to be  $3 \cdot 10^3$ . If ratios this high are encountered at the PPG it should be straightforward to determine the concentrations of the isotopes by gamma-spectrometry. Measurements of concentrations relative to  $^{207}\text{Bi}$  could help to resolve how and when the isotopes were produced. Surface sediments from “Bravo crater” at Bikini Atoll were previously shown to contain relatively high levels of  $^{207}\text{Bi}$ .<sup>3,4,26</sup> It was assumed that measurable amount of  $^{208}\text{Bi}$  might also be associated with sediment from the crater. Two cores were collected from the northern rim inside Bravo Crater during 1998 for analysis by gamma-spectrometry. Several archived samples were also analyzed to provide additional data for  $^{207,208}\text{Bi}$ . A summary of  $^{207}\text{Bi}$  in the environment of the PPG is

provided with a discussion of the relative results for  $^{208}\text{Bi}$  and  $^{207}\text{Bi}$  determined in the sediment samples.

### Nuclear testing and detection of $^{207}\text{Bi}$ in the environment

Many of the early scenarios explaining production of  $^{207}\text{Bi}$  at the PPG indicate it was a product of many if not all the atmospheric tests. The nuclear yields for different series of tests conducted between 1946 and 1958 at Enewetak and Bikini atolls are shown in Table 1 along with inventories (decay corrected to 1/1/2000) for several long-lived radionuclides still remaining in the lagoon sediments.<sup>5</sup> The total yield<sup>27</sup> at Bikini exceeds the yield at Enewetak. It would be anticipated to find inventories in the lagoon sediments higher at Bikini atoll. Table 1 shows this is the case for the fission products,  $^{137}\text{Cs}$ ,  $^{155}\text{Eu}$ ; for  $^{241}\text{Am}$  and  $^{239+240}\text{Pu}$ ; and possibly for the activation product,  $^{60}\text{Co}$ . However, the opposite is found for  $^{207}\text{Bi}$  where larger amounts are found associated with the sediments at Enewetak. The levels of  $^{207}\text{Bi}$  are not proportional to yield and must have been produced during specific tests rather than from many or all the nuclear events. Confirmation of this statement is augmented in the discussion to follow.

Descriptions given to early nuclear weapons include the terms “clean” or “dirty”.<sup>23,24,27</sup> Weapon components used in early tests were usually configured with quantities of natural, depleted or enriched uranium or other potentially fissionable material.<sup>23</sup> The energetic neutrons ( $>1.5$  MeV) produced during fusion (or from fission of the primaries) were capable of inducing fission in  $^{238}\text{U}$  (or other fissionable material) leading to an increase in fission yield. Weapons so configured were known as “dirty” because the end result was a release to the environment of larger amounts of radioactive fission products. SUBLETTE,<sup>24</sup> among others, states that in a “clean” device the  $^{238}\text{U}$  is replaced with other non-fissionable material. In a “clean device” most of the energy is derived from fusion whereas in a “dirty” device nuclear fission accounts for the majority of the total yield. Table 2 shows all tests conducted in the different areas of Bikini atoll. Yields<sup>27</sup> are shown and a “clean-dirty” designation is indicated when provided.<sup>27</sup> The Bravo, Koon and Zuni tests were surface explosions from platforms on coral soil. The other tests (except for the two tests conducted in 1946) were fired on barges or landing crafts moored over different depths of water in craters or the lagoon. All “clean” devices were exploded after the 1954 “Castle” series.

**Table 1.** Nuclear testing at Enewetak and Bikini Atolls and inventories of several long-lived radionuclides in lagoon sediments

Year of test series	Number of test	Total yield, Mt	<sup>241</sup> Am	Inventories of <sup>207</sup> Bi	radionuclides <sup>137</sup> Cs	in lagoon * c o	sediments <sup>a</sup> <sup>155</sup> Eu	<sup>239+240</sup> Pu
<b>Enewetak Atoll</b>								
1948	3	0.1						
1951	4	0.4						
1952	2	10.9						
1954	1	1.7						
1956	11	2.6						
1958	22	16						
Totals	43	31.7	24.5	23.3	3.3	0.22	1.0	62
<b>Bikini Atoll</b>								
1946	2	0.05						
1954	5	46.5						
1956	6	18.3						
1958	10	12.0						
Totals	23	16.8	90	1.9	4.3	0.29	1.5	122

<sup>a</sup> Values from Ref. 5 given in **TBq** to a depth of 30 cm in the lagoon. Decay corrected to 1/1/2000.

**Table 2.** Names and yields of nuclear tests at Bikini Atoll listed relative to locations

Name of event	Date of event, UTC	Total yield, kt	Added notes	
Bravo	2/28/54	15000	No <sup>207</sup> Bi with fallout samples (see text)	
Romeo	3/26/54	11000		
<b>Fir</b>	5/11/58	1360	Clean	
Sycamore	5/31/58	92	Clean	
Aspen	6/14/58	319		
Cedar	7/2/58	220	Clean	
Poplar	7/12/58	9300	Clean	
Tewa	7/20/56	5000		
Union	4/25/54	6900		<sup>203</sup> Pb with cloud sample
Yankee	5/4/54	13500		16000 lbs of Pb with vehicle
Cherokee	5/20/56	3800	?	
<b>Flathead</b>	6/11/56	365		
Dakota	6/25/56	1100	?	
Navaho	7/10/56	4500	Clean	<sup>203</sup> Pb in cloud samples
Maple	6/10/58	213		
Redwood	6/27/58	412		
Able	6/30/46	21		
Baker	7/24/46	21		
Koon	4/6/54	110		
Zuni	5/27/56	3500	Clean	<sup>203</sup> Pb in cloud
Hickory	6/29/58	14		
Nutmeg	5/21/58	25		
Juniper	7/22/58	65		

A number of investigators insist that lead was the target material required for the production of <sup>207</sup>Bi through charged particle reactions. Different quantities of lead were known to have been associated with a number of early tests.<sup>28</sup> For example, the “Mike” test in 1952 had a total yield of 10.4 Mt and was the largest

conducted at Enewetak. This test had the potential to produce some of the <sup>207</sup>Bi that is still measurable (Table 1) in the environment of Enewetak. It met the suggested requirements of being a high yield thermonuclear test with quantities of lead.

Our laboratory recently accepted ownership of some marine and terrestrial samples from the PPG and other Marshall Island atolls collected during and after the years of nuclear testing. The samples had been in storage at the College of Fisheries, University of Washington, Seattle, WA. Analysis of the samples by gamma-spectrometry provides information regarding what long-lived, gamma-emitting, radionuclides existed in the environment at the times of collection, and helps to establish when  $^{207}\text{Bi}$  first appeared in the environment. Samples were counted between October and December 1999. At Enewetak the samples were obtained from the reef, islands and the lagoon at a distance no greater than 5 miles from the crater resulting from the 10.4 Mt Mike explosion.  $^{207}\text{Bi}$  was not found in any aquatic or terrestrial sample collected from this region before 6/19/58. The fission product,  $^{137}\text{Cs}$ , was present in all samples. The absence of  $^{207}\text{Bi}$  from the samples collected prior to 1958 is noteworthy. If the Mike test or, for that matter, any other tests in the pre-1958 series produced  $^{207}\text{Bi}$ , the radionuclide would have been detected in one or more of the samples collected before early 1958. Some additional samples from the remainder of the atoll were also analyzed. None were found to contain any quantity of  $^{207}\text{Bi}$ . It is concluded that  $^{207}\text{Bi}$  could not have originated in the "Mike" test, with its quantities of lead, or any of the other tests (some associated with quantities of lead)<sup>28</sup> held at Enewetak before 1958. Therefore, it is not merely the association of a target material with a device but the manner in which it is configured relative to the irradiating particles that is important for generating many of the activated products.

A comparable number of archived samples from Bikini were also analyzed by gamma-spectrometry. There was no indication of  $^{207}\text{Bi}$  in any type of sample collected from the environment prior to September 1956. It was then measured in nearly all types of environmental samples from the atoll during the ensuing years. Recall that  $^{207}\text{Bi}$  was not detected anywhere in the Enewetak environment until late 1958. Therefore, the 1956 test debris containing  $^{207}\text{Bi}$  from Bikini Atoll did not contaminate Enewetak, 305 km to the west. The  $^{207}\text{Bi}$  produced at the PPG was not widespread throughout the equatorial Pacific or, for that matter, the global environment.

$^{203}\text{Pb}$  was detected in cloud samples from the Zuni, Tewa and Navaho tests.<sup>28</sup> Zuni and Navaho were "clean" devices (Table 2). Tewa was essentially a "dirty" version of the Zuni test. The  $^{203}\text{Pb}$  can only result from the  $^{204}\text{Pb}(n,2n)$  reaction that has a threshold energy of 8.4 MeV. Consequently, stable lead must have been configured with the 3 devices so that activation of the material was accomplished with high-energy neutrons.

Any one of these 3 tests could have generated the  $^{207}\text{Bi}$  detected in the 1956 library samples from Bikini provided that sufficiently energetic charged particles were available to interact and activate the lead. There is no available information to show if bismuth or a bismuth-lead alloy was *also* a component associated with these devices.

Table 3 lists previously unpublished concentrations of some radionuclides detected in sections of two sediment cores sampled in 1972 from Bravo crater. The Bravo event was the largest thermonuclear test recorded at the PPG. It produced a crater in the northwest reef over 1800 m in diameter. Test Bravo, however, was not responsible for producing the  $^{207}\text{Bi}$  and other radionuclides shown in Table 3. Debris originating from the Bravo event contaminated the Japanese fishing vessel, Fukukyu Maru that was located in the equatorial Pacific, miles to east of Bikini. A sample of this fallout material was provided to us for analysis. SHIMIZU<sup>29</sup> analyzed aliquots of the fallout debris by gamma-spectrometry in 1985 and we counted our fraction 10 years later. The man-made gamma-emitting radionuclides detected in the samples at both laboratories included only  $^{241}\text{Am}$ ,  $^{155}\text{Eu}$ ,  $^{137}\text{Cs}$  and  $^{60}\text{Co}$  but no  $^{207}\text{Bi}$ . The tests conducted in 1954 accounted for 61% of the total yield at Bikini. The absence of  $^{207}\text{Bi}$  from any environmental samples at Bikini collected during 1954, 1955 or early 1956 confirm the results from the Fukukyu Maru analysis and also rule out any possible production of  $^{207}\text{Bi}$  by the other high yield tests in the 1954 "Castle" series.

Figure 1 and Table 2 show that Poplar was the last of the 7 events exploded in Bravo crater. It was also the last large (8.8 Mt) "clean" test in the 1958 series. The device underwent 32 design modifications before being fired.<sup>23</sup> In all probability, Poplar was responsible for producing the  $^{207}\text{Bi}$  (and other radionuclides) now found in the surface sediments of the crater. It has not been determined if Poplar contained critical components made of either lead or bismuth. However, some high  $Z$  unfissionable material must have been used in the device. The Poplar design was similar to that of the 2 Mt (estimated 90% fusion yield) "clean" Pine event fired at Enewetak on 26 July 1958.<sup>27</sup> The lagoon sediment in the local area of the Pine test site contains the highest levels of  $^{207}\text{Bi}$  now found in Enewetak lagoon.  $^{207}\text{Bi}$  was not detected in the archived samples prior to the Pine test. This test possibly could have generated the majority of  $^{207}\text{Bi}$  currently detected in the Enewetak atoll environment. No specific information is available on the use of either lead or bismuth with this device. Its high fusion-low fission yield indicates some high  $Z$  material(s) was used in place of  $^{238}\text{U}$ .

Table 3. Bikini Bravo crater core samples collected 13 November 1972

ID#	Depth increment, cm	Dry wt, g	$^{241}\text{Am}$	$^{155}\text{Eu}$	Radionuclide concentrations, $\text{Bq}\cdot\text{kg}^{-1}$ dry weight						$^{101}\text{Rh}$	$^{101}\text{Rh};^{102}\text{Rh}$
			$^{60}\text{Co}$	$^{137}\text{Cs}$	$^{207}\text{Bi}$	$^{102m}\text{Rh}$	$^{125}\text{Sb}$					
Core Station C2 <sup>a</sup>												
1	0-6	86	1541	1619	1085	272	2330	61	89	10.4	0.17	
2	6-12	155	1567	1833	1126	313	2896	73	103	10.4	0.14	
3	12-18	188	1622	1885	5330	335	7481	101	678	20.7	0.20	
4	18-24	183	1667	1911	6589	361	8556	83	1030	18.1	0.22	
5	24-30	177	1544	1793	6163	336	8074	104	952	15.9	0.15	
6	30-36	192	1570	1830	6285	357	8185	87	985	19.3	0.22	
7	36-42	187	1619	1870	6481	374	8370	91	941	17.0	0.19	
8	42-48	195	1619	1900	6578	378	8556	90	1089	16.3	0.18	
9	48-54	182	1796	2022	7137	415	9296	115	1041	18.9	0.16	
10	54-60	229	1533	1637	5859	374	7519	82	785	17.0	0.21	
11	60-66	201	1563	1781	6311	400	8111	94	974	17.8	0.19	
12	66-72	195	1622	1952	6774	404	8704	73	1022	18.5	0.25	
Core Station C3 <sup>b</sup>												
1	0-6	213	1356	1599	2156	345	4515	88	200	12.2	0.14	
2	6-12	196	1130	1174	3470	248	4559	47	426	11.5	0.24	
3	12-18	236	548	489	1319	227	933	20	178	4.4	0.23	
4	18-24	237	507	430	1144	228	689	14	170	4.4	0.32	
5	24-30	240	511	404	1059	223	581	17	133	2.6	0.15	
6	30-36	235	526	430	1185	237	563	15	163	2.9	0.19	
7	35-42	232	611	448	1126	232	489	13	167	3.1	0.24	
8	42-48	238	967	644	1330	259	767	20	196	4.1	0.20	

<sup>a</sup> Water depth 47m; 11°41'28"N, 165°16'37"E. Counting errors less than 2%. Concentrations on date of collection.

<sup>b</sup> Water depth 47.4m; 11°41'25"N, 165°16'10"E. Counting errors less than 3%. Concentrations on date of collection.

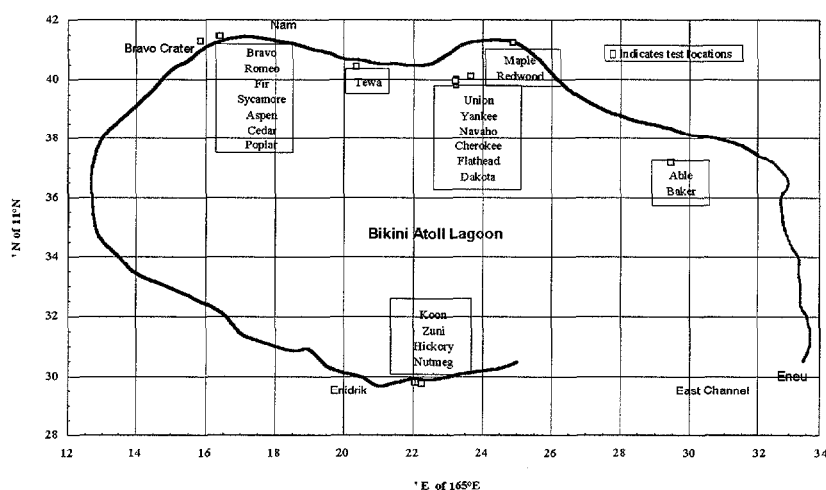


Fig. 1. Locations of all nuclear tests at Bikini Atoll listed in Table 2

### Processing of field samples, gamma-spectrometry and results for $^{208}\text{Bi}$ and $^{207}\text{Bi}$

The cores were frozen and returned to the laboratory for sectioning into 4-cm increments. Samples were dried, pulverized, blended and aliquots of the carbonate sediment packaged into counting vials. Full cylindrical vials were 4 cm high and 9.62 cm<sup>2</sup> in area and contained approximately 42 g of dry sediment. A single high purity

Ge detector with a Be window was used to count each of the sectioned samples for periods of 1 to 3 days. The activity under the 2614 keV peak consisted of 3 components. First was the natural background contribution from  $^{208}\text{Tl}$  (from natural  $^{232}\text{Th}$  daughters). The second component was the activity resulting from decay of  $^{208}\text{Bi}$  in the sample and the third was any  $^{208}\text{Tl}$  in the sample supported by decay from  $^{232}\text{Th}$  daughters.

There is another abundant gamma-ray from  $^{208}\text{Tl}$ -decay easily identified and free of interference at 584 keV. A  $^{232}\text{Th}$  standard was used to determine the relative activity of the 584 and 2614 keV photopeaks. The ratio was used in conjunction with the measured activity at 584 keV to estimate the contribution of  $^{208}\text{Tl}$  to the 2614 keV peak in the sample. The counting data were then corrected for background and any contribution for  $^{208}\text{Tl}$  in the sample. The activity remaining after subtraction was assumed to be  $^{208}\text{Bi}$ . Concentrations of  $^{208}\text{Bi}$  are shown in Table 4. Large errors were associated with the concentrations in many sections and resulted from the low activity and corrections for contributions of background and any supported  $^{208}\text{Tl}$ .

A few of the 1998 sediment samples were dissolved and bismuth was separated by solvent extraction into a chloroform solution containing diethylammonium-diethyldithiocarbamate. The bismuth was precipitated as  $\text{BiOCl}$  and prepared for gamma-counting  $^{207,208}\text{Bi}$ . There was no evidence of the 584 keV gamma-peak from  $^{208}\text{Tl}$  in these samples so only background contributed to the activity of the 2614 keV peak.

The presence of excess activity in the 2614 keV peak in the separated bismuth samples confirmed that the activity determined by difference in the chemically unprocessed samples was  $^{208}\text{Bi}$ . In other processed samples no  $^{208}\text{Bi}$  activity was found. This finding confirmed that the level initially measured in the unprocessed sample was indeed below detection. Several archived samples from Bravo Crater with higher levels of  $^{207}\text{Bi}$  were also prepared for counting on the same gamma detector. Table 4 shows the results for all measured activities. Crater samples that were gamma-counted in late 1999-early 2000 revealed only the presence of different amounts of the man-made radionuclides,  $^{241}\text{Am}$ ,  $^{155}\text{Eu}$ ,  $^{60}\text{Co}$ ,  $^{137}\text{Cs}$  and  $^{207}\text{Bi}$ .  $^{125}\text{Sb}$  and  $^{101,102\text{m}}\text{Rh}$ , identified in the 1972 crater sediments and shown in Table 3, had decayed and were below detection limits by 1999. Only results for the bismuth isotopes and  $^{137}\text{Cs}$  (for information) are shown in Table 4. All values in Table 4 have been decay corrected to 7/12/58 ( $t_0$ ), the date of the Poplar test in Bravo crater. The atom ratio of  $^{208}\text{Bi}$  to  $^{207}\text{Bi}$  shown in the last column of Table 4 will hereafter be referred to as  $R$ .

Table 4.  $^{207,208}\text{Bi}$  gamma counting results, activity and atom ratios

ID	Section depth, cm	$^{137}\text{Cs}$ , <sup>a</sup> $\text{Bq}\cdot\text{kg}^{-1}$	Error, %	$^{207}\text{Bi}$ , <sup>a</sup> $\text{Bq}\cdot\text{kg}^{-1}$	Error, %	$^{208}\text{Bi}$ , $\text{Bq}\cdot\text{kg}^{-1}$	Error, %	$^{208}\text{Bi}:^{207}\text{Bi}$ activity ratio	$\pm\text{Error}$	$^{208}\text{Bi}:^{207}\text{Bi}$ atom ratio ( $R$ )
Bravo crater sediment collected 1 1/98										
Core 1										
2880	0-4	298	0.9	332	2	1.12	40	0.003	0.001	38
replicate	0-4	301	0.5	341	1	1.12	45	0.003	0.001	37
2880R <sup>b</sup>	0-4			340	1	0.80	20	0.002	0.000	27
2881	4-8	197	1.2	127	2	0.30	100			
2881R <sup>b</sup>	4-8			133	1	co.2				
2882	8-12	186	1.4	100	2	0.16	100			
2882R <sup>b</sup>	8-12			110	2	0.05	120			
2883	12-16	158	1	97	1.4	-0.20	400			
core 2										
2889	0-4	171	1.3	91	1.5	1.92	40	0.021	0.008	240
2890	4-8	139	1.7	60	2.4	0.71	46	0.012	0.005	135
2891	8-12	143	1.3	52	2.3	1.13	46	0.022	0.010	248
2892	12-16	141	1.4	53	2.3	0.96	68	0.01s	0.012	207
2893	16-20	152	1.3	57	2.3	0.99	75	0.017	0.012	198
2896	28-32	121	2	62	3	0.70	75	0.012	0.009	135
Bravo crater sediment from 42 m collected 5/6/72										
surface		414	1	2500	1	3.40	15	0.0014	0.0002	16
Bravo crater sediment Sta C3+ collected 1 1/7/72										
surface		380	2	2800	1	2.90	20	0.0010	0.0002	12
Crater North shallow rim collected 5/6/72										
surface		3900	0.5	16	4	-0.10	150			

<sup>a</sup> Data corrected for decay from counting date to 7/12/58.

Also present in gamma-spectrum:  $^{155}\text{Eu}$ ,  $^{241}\text{Am}$  and  $^{60}\text{Co}$ . Concentrations calculated but are not shown.

<sup>b</sup> Radiochemically separated bismuth

## Discussion

Small amounts of  $^{208}\text{Bi}$  were measured in some of the sediments from Bravo Crater. The quantities detected are attributable to the 7/12/58 Poplar event. The lowest  $^{208}\text{Bi}$ :  $^{207}\text{Bi}$  activity ratio determined at  $t_0$  with some degree of confidence is about 0.001. The respective atomic ratio ( $R$ ) would be 12. Lower  $R$  values are possible since the two sigma error is about  $\pm 5$ .  $R$  values associated with core 2 samples have large associated errors. However, as a group, these samples suggest higher values of  $R$  are associated with some of the crater sediments. It appears that  $^{207,208}\text{Bi}$  isotopes were not uniformly distributed over the surface area of the crater. The isotopes could not have been well mixed with the debris resulting from the explosion. No  $R$  value as high as  $3 \cdot 10^3$ , reported by SHINOHARA et al.,<sup>21</sup> was measured in the crater samples.

### Scenarios for the $^{209}\text{Bi}(n,xn)^{207,208}\text{Bi}$ reaction

The production of activities using the standard activation Eq. (1) is reduced to Eq. (2) in a thermonuclear explosion since  $t$  is short compared to  $T_{1/2}$ .  $A$ , the activity, is equal to  $N\lambda$  where  $N$  represents the number of atoms of the respective radionuclide. The definitions of  $\sigma$ ,  $\phi$  and  $n$  are the usual cross section, flux and target atoms, respectively:

$$A = \sigma \phi n (1 - e^{-\lambda t}) \quad (1)$$

$$N = \sigma \phi n t \quad (2)$$

The relationship holds for production of either  $^{208}\text{Bi}$  or  $^{207}\text{Bi}$  by activation of  $^{209}\text{Bi}$  with fast neutrons. The  $R$  value is given by Eq. (3). At the same activation energy, the values for  $\phi$ ,  $n$ , and  $t$  are identical for both radionuclides and cancel.  $R$  is then simply the ratio of cross sections at any neutron energy:

$$N_1:N_2 = \sigma_1:\sigma_2 = R \quad (3)$$

Files of nuclear data given in JENDL (Japanese data base) and JEF (European data base) provide interpretative cross section data for  $^{209}\text{Bi}$  ( $n,xn$ ) reactions.<sup>1</sup> At 15 MeV the mean ratio computed from both data bases is  $390 \pm 50$  while at 16 MeV the value falls to 28.5.<sup>1</sup> Over this energy range the formation cross section for  $^{208}\text{Bi}$  is relatively constant. The change in  $R$  is due to the increasing formation cross section of  $^{207}\text{Bi}$  as the activation energy increases. Therefore, relatively less  $^{207}\text{Bi}$  should be associated with samples having high  $R$  values which is the situation found with the different crater sediments. The  $^{209}\text{Bi}(n,2n)$  and  $^{209}\text{Bi}(n,3n)$  reaction cross sections ( $\sim 1.2$  b) are comparable at about 19.5 MeV.<sup>1,30</sup> At this energy the

$R$  value is 1 (activity ratio  $\sim 9 \cdot 10^{-5}$ ). With the levels of  $^{207}\text{Bi}$  encountered in the sediment samples, there would be little chance in determining an  $R$  of 1, or much less than 5, by gamma-spectrometry. Energetic neutrons could generate  $R$  values ranging from the 100's to less than 10. Thus finding different  $R$  values in the environment would be predicable if the isotopes had been a product of ( $n,xn$ ) reactions on stable bismuth.

Table 3 shows that  $^{101}\text{Rh}$  ( $T_{1/2} = 3.3$  y) and  $^{102m}\text{Rh}$  ( $T_{1/2} = 2.9$  y) were also present in the 1972 sediments from Bravo Crater. These rhodium isotopes had decayed and were below detection limits in the 1998 core samples. The mean activity ratio of  $^{101}\text{Rh}$ : $^{102m}\text{Rh}$  during Nov. 1972 in core C2 to a depth of 72 cm was  $0.19 \pm 0.03$  and in C3 the mean value was  $0.21 \pm 0.06$ . The activity ratio at  $t_0$  would have been  $\sim 0.13 \pm 0.02$  and the atom ratio would have been  $0.14 \pm 0.02$ . The independent fission yield of the two radionuclides is very small. Therefore,  $^{101,102m}\text{Rh}$  most likely resulted from  $^{103}\text{Rh}$  ( $n,xn$ ) reactions in the device. Reports identify  $^{102m}\text{Rh}$  in early debris from many of the 1958 (Hardtack series) tests including Poplar.<sup>28</sup> However, these same reports make no mention of  $^{101}\text{Rh}$  which was possibly masked from detection by other shorter lived isotopes. The thresholds for the ( $n,3n$ )  $^{101}\text{Rh}$  and the ( $n,2n$ )  $^{102m}\text{Rh}$  reactions are 16.8 and 9.3 MeV,<sup>1</sup> respectively. Neutrons with energies well over 14.1 MeV must have been generated in the Poplar explosion to interact with stable rhodium and produce  $^{101}\text{Rh}$ . Calculated neutron cross section data is available for the ( $n,2n$ ) and ( $n,3n$ ) reactions in the JENDL data base.<sup>1</sup> From Eq. (3) the ratio of the rhodium isotope cross sections at any neutron energy is proportional to the atom ratio at the time of the explosion. At neutron energies of 18, 19 and 20 MeV the atom ratio for  $^{101}\text{Rh}$ :  $^{102m}\text{Rh}$  is computed to be 0.042, 0.214, and 0.458, respectively. The measured atom ratio of 0.14 suggests the isotopes were formed during activation of stable rhodium with 18–19 MeV neutrons. This energy range is well above the threshold energies for forming not only  $^{101,102m}\text{Rh}$  isotopes but  $^{207}\text{Bi}$  as well. Therefore, sufficiently energetic neutrons would have been available to produce  $^{207}\text{Bi}$  by activation of  $^{209}\text{Bi}$  if it was associated with the Poplar device. Using the JENDL and JEF cross sections estimates<sup>1</sup> previously discussed, the activation of  $^{209}\text{Bi}$  with 18 MeV neutrons would generate an  $R$  value of 2.8 while at 19 MeV the  $R$  value would be 1.5. As previously acknowledged, activation with neutron having energies less than 17 MeV would result in higher  $R$  values with lower  $^{207}\text{Bi}$  concentrations. Therefore, all the measured  $R$  values and concentrations in the sediment samples are consistent with a scenario involving the activation of  $^{209}\text{Bi}$  with a spectrum of fast neutrons having energies between 14 and 19 MeV.

*Scenarios for charged particle activation*

BELL<sup>31</sup> indicates that any  $^{238}\text{U}$ , if present in a thermonuclear device, exposed to an intense fast neutron flux will undergo fission, and (n,2n), (n,3n) and (n,p) reactions. In addition, collisions of the neutrons with  $^2\text{H}$  or  $^3\text{H}$  will involve energy transfer leading to the possibility that the energetic particles can produce (d, $\gamma$ ), (d,n), (d,3n), (t, $\gamma$ ), (t,n), (t,2n) and (t,3n) reactions on uranium. In collisions with a 14.1 MeV neutron, for example, a deuteron can acquire up to 12.5 MeV, a triton (and  $^3\text{He}$ ) up to 10.6 MeV and a proton up to 14.1 MeV. Fusion of these energetic deuterons and tritons will in turn also produce neutrons with energies much higher than 14.1 MeV. The principal source of Np production in a thermonuclear device is believed to arise from a fast deuteron reaction with uranium.<sup>31</sup> Many of these energetic particles could also react with any lead associated with a device and produce  $^{207,208}\text{Bi}$ . Reactions with threshold energies below 6 MeV<sup>1</sup> include  $^{206}\text{Pb}(\text{p},\gamma;\text{d},\text{n};\text{t},2\text{n})$  and  $^{207}\text{Pb}(\text{p},\text{n};\text{d},2\text{n})$  leading to  $^{207}\text{Bi}$  and  $^{206}\text{Pb}(\text{d},\gamma;\text{t},\text{n})$ ,  $^{207}\text{Pb}(\text{p},\gamma;\text{d},\text{n};\text{t},2\text{n})$  and  $^{208}\text{Pb}(\text{p},\text{n};\text{d},2\text{n})$  leading to  $^{208}\text{Bi}$ . Another reaction to consider with  $^{209}\text{Bi}$  is ( $^3\text{He}$ , n). This will produce  $^{211}\text{At}$  which undergoes decay to  $^{207}\text{Bi}$ . Neutron loss is the dominate mode of compound nucleus decay for protons and probably other charged particle reactions with energies near  $8 \pm 3$  MeV.<sup>32-34</sup> Therefore, reactions involving the emission of most other particles (p,  $\alpha$ , 3H, etc) can be neglected.

If natural lead alone were the target material most of the above mentioned charged particles reactions would produce the bismuth isotopes with a low  $R$  value, probably no greater than 2–3 (activity ratio of  $\sim 0.0002$ ). Table 5 shows a listing of  $Q$  values for (p,n), (d,n), (d,2n), (t,2n) (p, $\gamma$ ) reactions on lead leading to formation of  $^{207}\text{Bi}$  and  $^{208}\text{Bi}$ .<sup>1</sup> Reactions are grouped in pairs to show the similarity in  $Q$  for the same charged particle reaction with the different lead isotopes (208, 207, 206). The  $Q$  values for the same reactions are seen to differ by no more than a few tenths of a MeV. It is further assumed, for example, that the  $\sigma(\text{p},\text{n})$  cross section for either  $^{207}\text{Pb}$  or  $^{208}\text{Pb}$  at any proton energy would be essentially the same, differing by no more than the difference noted between the  $Q$  values. In other words the excitation function for the  $^{208}\text{Pb}(\text{p},\text{n})$  reaction forming  $^{208}\text{Bi}$  (threshold = 3.7 MeV) would have the same shape and values as the excitation function for the  $^{207}\text{Pb}(\text{p},\text{n})^{207}\text{Bi}$  (threshold = 3.2 MeV) reaction. If only  $^{207,208}\text{Pb}(\text{p},\text{n})$  reactions produced the bismuth isotopes, the  $R$  value in Eq. (3) would reduce to the ratio of the natural abundance of  $^{208}\text{Pb}$  to  $^{207}\text{Pb}$  or 2.4 (52.4 divided by 22.1). The resulting activity ratio of  $^{208}\text{Bi}$ :  $^{207}\text{Bi}$  would be 0.0002. The same  $R$  value would result if the

$^{207,208}\text{Pb}(\text{d},2\text{n})$  or  $^{207,208}\text{Pb}(\text{t},2\text{n})$  were the principal reactions leading to  $^{207}\text{Bi}$  and  $^{208}\text{Bi}$ . The  $R$  value would not exceed 2.4 if the three reactions together contributed to the formation of  $^{207,208}\text{Bi}$ . An  $R$  value of 0.92 (the abundance ratio of natural  $^{207}\text{Pb}$  to  $^{206}\text{Pb}$ ) would have been found in the samples (but not measured by gamma spectrometry) if the  $^{206,207}\text{Pb}(\text{d},\text{n})$ , (t,2n), (p, $\gamma$ ) and ( $^3\text{He}$ ,d) reactions were responsible for all of the  $^{207,208}\text{Bi}$  formed in the Poplar event. In order to increase  $R$  to a value near or greater than 10, reactions are necessary that will increase the relative amount of  $^{208}\text{Bi}$  formed. Among the reactions to consider that result in only the production of  $^{208}\text{Bi}$  are  $^{206}\text{Pb}(\text{t},\text{n})$ , ( $^3\text{He}$ ,p) or (d, $\gamma$ ). These explanations regarding production of the bismuth isotopes from the different lead isotopes are speculative. However, an  $R$  value of  $\sim 10$  or greater would require that the charged particle cross sections with the lead isotopes differ by at least a factor of 5–10 at any irradiation energy. It seems unlikely that favorable reactions, for example, such as  $^{207}\text{Pb}(\text{d},\text{n})^{208}\text{Bi}$  and  $^{206}\text{Pb}(\text{d},\text{n})^{207}\text{Bi}$  at the same deuteron energy would have cross sections differing by a factor of 10. Lead could be the principal target material for the bismuth isotopes in a device but  $R$  values generated from any of the mentioned charged particle reactions would not be distinguished from  $R$  produced through  $^{209}\text{Bi}(\text{n},\text{xn})$  reactions with neutrons having energies greater than 18 MeV. Another consideration is to have lead present in conjunction with bismuth as an alloy in a device as suggested by SUBLETTE.<sup>24</sup> Activation of both materials could generate the relative amounts of the  $^{207,208}\text{Bi}$  measured in the samples. It would only require neutrons with energies between 7.5 and 14.1 MeV to interact with stable  $^{209}\text{Bi}$  and form some fraction of the  $^{208}\text{Bi}$  by an (n,2n) reaction. Any of the mentioned charged particle reacting on lead should generate quantities of both  $^{207}\text{Bi}$  and  $^{208}\text{Bi}$ . The supplemental quantity of  $^{208}\text{Bi}$  will increase the  $R$  value resulting from lead activation alone.

Determinations of  $R$  that are less than 10 in any environmental sample are extremely difficult by gamma-spectrometry. In this study it was not possible to confidently determine levels of  $^{208}\text{Bi}$  at levels below  $0.3 \text{ Bq}\cdot\text{kg}^{-1}$  by gamma-spectrometry with the experimental design. Accurate determination of  $^{208}\text{Bi}$  at a level below  $0.3 \text{ Bq}\cdot\text{kg}^{-1}$  would be necessary to arrive at accurate  $R$  values less than 10 with the range of concentrations for  $^{207}\text{Bi}$  encountered in the samples. It is of course possible to change experimental conditions for gamma analysis, i.e., directly count larger samples, find material with higher levels of  $^{207}\text{Bi}$ , count under conditions of lower background, and separate bismuth from larger samples. However, if more reliable data are required, the bismuth isotope ratios are better determined by mass spectrometry.



Table 5. Q-values for some different charged particle reactions leading to formation of  $^{207}\text{Bi}$  and  $^{208}\text{Bi}$  from natural lead isotope?

Reaction	Q-values MeV	Lead isotope abundance, %
$^{208}\text{Pb}(^3\text{He}, ^3\text{H})^{208}\text{Bi}$	-2.89	52.4
$^{207}\text{Pb}(^3\text{He}, ^3\text{H})^{207}\text{Bi}$	-2.41	22.1
$^{208}\text{Pb}(\text{p}, \text{n})^{208}\text{Bi}$	-3.66	52.4
$^{207}\text{Pb}(\text{p}, \text{n})^{207}\text{Bi}$	-3.18	22.1
$^{208}\text{Pb}(^2\text{H}, 2\text{n})^{208}\text{Bi}$	-5.89	52.4
$^{207}\text{Pb}(^2\text{H}, 2\text{n})^{207}\text{Bi}$	-5.40	22.1
$^{207}\text{Pb}(\text{p}, \gamma)^{208}\text{Bi}$	3.71	22.1
$^{206}\text{Pb}(\text{p}, \gamma)^{207}\text{Bi}$	3.56	24.1
$^{207}\text{Pb}(\text{d}, \text{n})^{208}\text{Bi}$	1.48	22.1
$^{206}\text{Pb}(\text{d}, \text{n})^{207}\text{Bi}$	1.33	24.1
$^{207}\text{Pb}(^3\text{He}, ^2\text{H})^{208}\text{Bi}$	-1.74	22.1
$^{206}\text{Pb}(^3\text{He}, ^2\text{H})^{207}\text{Bi}$	-1.94	24.1
$^{207}\text{Pb}(^3\text{H}, 2\text{n})^{208}\text{Bi}$	-4.78	22.1
$^{206}\text{Pb}(^3\text{H}, 2\text{n})^{207}\text{Bi}$	-4.92	24.1
$^{206}\text{Pb}(\text{p}, \gamma)^{208}\text{Bi}$	8.22	24.1
$^{206}\text{Pb}(^3\text{He}, \text{p})^{208}\text{Bi}$	2.73	24.1
$^{206}\text{Pb}(^3\text{H}, \text{n})^{208}\text{Bi}$	1.96	24.1

<sup>a</sup> Q-values are from Ref. 1.

Without any additional knowledge of the components associated with the Poplar test it is impossible to assess if only lead or bismuth or an alloy was activated to produce the  $^{207,208}\text{Bi}$  measured in the crater sediment samples. However, finding values of  $R$  that are  $>10$  suggest that a fraction of the  $^{207,208}\text{Bi}$  formed was by neutron activation of stable bismuth in the device.

### Conclusions

$^{207}\text{Bi}$  is found widely distributed in the environments of both Bikini and Enewetak Atolls. The radionuclide was produced in only a few “clean” tests conducted at the PPG after the 1954 test series. At Enewetak  $^{207}\text{Bi}$  was first detected in samples collected in late 1958 while at Bikini it was found in samples collected during and after the 1956 test series. In addition to  $^{207}\text{Bi}$ , small concentrations of  $^{208}\text{Bi}$  have been measured in selected sediment samples from a nuclear crater in Bikini atoll. The contamination in the surface sediments is believed to have originated from the Poplar event on 7/12/1958. Atom ratios ( $R$  values) of  $^{208}\text{Bi}:^{207}\text{Bi}$  in the crater sediments range from  $\sim 12$  to over 200. It is argued that the highest ratios are only possible if  $^{209}\text{Bi}$  was a component or alloyed with lead in the device. Both isotopes could result from  $^{209}\text{Bi}(\text{n}, \text{xn})$  reactions with neutrons with energies in excess of 14.1 MeV. Such energetic neutrons were required in the Poplar test to account for the presence of  $^{101}\text{Rh}$  in the sediments. If

lead and bismuth were associated with the device, two forms of activation are proposed for producing the relative amounts of  $^{207,208}\text{Bi}$  found in the sediments. Activation of lead by any number of charged particle reactions will produce both bismuth isotopes with a low atom ratio ( $<3$ ). Activation of bismuth with fast neutrons having energies between 7.5 and 14.1 MeV would lead to the formation of  $^{208}\text{Bi}$  only. Added together the bismuth isotopes from both target materials would result in an  $R$  value greater than 10. Lead was a known component in some tests but was not properly configured to generate any  $^{207}\text{Bi}$  by charged particle activation. No information can be found in the literature to assess what tests conducted at the PPG contained stable bismuth in any form. Identities of these tests would greatly help in interpreting the reaction mechanisms leading to the formation of  $^{207}\text{Bi}$  and  $^{208}\text{Bi}$ .

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